

Argonne National Laboratory

NUCLEAR-FUEL-ELEMENT LOADING BY VIBRATORY COMPACTION-- URANIUM-PLUTONIUM CARBIDE SPECIMENS FOR EBR-II IRRADIATION

by

J. E. Ayer, F. E. Soppet,
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ABSTRACT

This report presents the results of fabrication by vibratory compaction of the first of a series of fuel specimens. The uranium-plutonium carbide specimens made for this study were jacketed in eight different alloys. The fuel rods were manufactured by an infiltration technique developed at ANL. The loading conditions were derived from basic information gathered on a model system.^{1,2}

Eleven mixed-carbide rods were made, ten of which were loaded to 80% theoretical density (TD) with 80 w/o uranium carbide and 20 w/o plutonium carbide. The fuel bodies were low in both density and plutonium carbide in the upper 2 in. of the column. One mixed-carbide specimen, compacted to 83.8% TD and containing 19.3% plutonium carbide, was more uniform in density and plutonium distribution than the ten based upon the model system.

Five fuel specimens containing solid-solution uranium-plutonium carbide were fabricated by the infiltration technique to the highest density practicably attainable. The five specimens showed uniform density and fuel distribution. The density of the fuel columns was $84.2 \pm 1.3\%$ of TD attained with 97.1% TD particles compacted into 6.5-mm-ID jackets.

INTRODUCTION

Since 1963 a program directed toward gaining a facility for fuel-element manufacture by vibratory-compaction techniques has been underway at ANL. This program, coupled with a capacity for the synthesis of plutonium carbide,³ has developed into a modest capability for carbide compaction. In December 1964 a request was made for sixteen vibratory-compacted carbide fuel rods for irradiation in EBR-II. The fabrication of

these sixteen specimens, the first full-scale vibratory-compacted elements made at ANL, is the subject of this report.

FUEL-ELEMENT DESCRIPTION

The requested fuel specimen consisted of a right-circular cylindrical metal jacket with welded end plugs, fuel column, and retainer. A drawing of the fuel specimen and its pertinent dimensions is shown in Fig. 1.

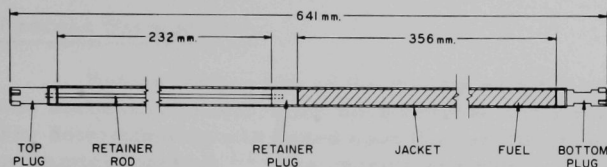


Fig. 1
EBR-II Vibratorily-compacted
Carbide Irradiation Specimen

One of the purposes of the irradiation experiment was to determine the extent of reaction of carbide fuels with various jacket materials in a reactor environment. Accordingly, a series of jacket-fuel combinations were provided. Table I is a listing of the samples manufactured. The term UC-PuC in the table represents a physical mixture of UC and PuC; (U·Pu)C is the designation employed for a solid solution containing UC and PuC.

TABLE I. Types of Samples Manufactured

Number of Specimens	Fuel Form	Jacket Material
4	UC-PuC	Hastelloy-X
3	UC-PuC	Nb-1w/oZr
2	UC-PuC	Ta-10w/oW
1	UC-PuC	316 Stainless
1	(U·Pu)C	304 Stainless
2	(U·Pu)C	Nb-1w/oZr
1	(U·Pu)C	V-20w/oTi
1	(U·Pu)C	Vanadium
1	(U·Pu)C	W-coated Hastelloy-X

The fuel body, specified as vibratorily compacted, was required to be 20% PuC and 80% UC. A uniform distribution of PuC in the mixed UC-PuC was desired. The density of the mixed-carbide system was specified to be 80% TD. Since the distribution and concentration of PuC in a solid solution with UC is uniform, the only requirement for the solid-solution fuel body was that its density be uniform. For solid-solution specimens, as high a density as might be practicably attained was allowed.

The finished fuel specimen was accepted when it was leak-tight and free of alpha contamination. A rod was sufficiently clean when it showed less than 400 dpm direct count and zero wipe count using a proportional alpha counter. Leaktight was construed as loss of helium from the jacket, under a one-kg/cm² differential, at a rate less than 2×10^{-6} std cc/sec as determined by a helium mass-spectrometer leak detector.

PROCEDURE FOR MANUFACTURING FUEL RODS

Process Development

Before loading any of the jackets with the two carbide materials, it was necessary to determine the quantities and sizes of grains to be loaded. The determination was based upon the results of newly developed information on particle packing.^{1,2} The packing process to be used consisted of an infiltration technique.⁴ Experimentation with malleable, nonfissile materials compacted into rigid tubes indicated that the maximum packing fraction for a two-component system of angular particles in a small-diameter (6.5-mm) tube would be about 80%. Since the particle density of the carbides was about 97.5%, such a packing fraction would yield a fuel density of only 78% TD.

An obvious solution to the problem of raising the packing density would, in a model system, be the use of three components. However, we had found that efforts to produce uniform density in ternary systems of angular particles were singularly unsuccessful. An alternative would be to use the popular procedure of blending three components and pouring them into the jackets while vibrating.^{5,6} This solution was rejected because of nonuniform density and particle distribution resulting from the rapid settling of the finest fraction and subsequent levitation of the coarser particles.

The procedures to yield an acceptable product involved compromise between the two solutions suggested above. A three-component system was devised in which settling and levitation were prevented, and the infiltration of the finest fraction came about with little difficulty. To accomplish this a blend of two sizes of components was made. The coarse fraction size was chosen to yield the maximum packing efficiency of angular particles in a 6.5-mm tube according to the equation²

$$Pe_1 = 0.635 - \frac{32.0}{d_1} - 0.072 e^{-0.207D/d_1},$$

where

Pe_1 is the packing efficiency of the coarse component;

d_1 is the particle dimension (in microns);

D is the inside diameter of the tube (in microns).

The optimum size for d_1 was found to be 1.2 mm. The determination was made by taking the derivative of Pe_1 with respect to d_1 in the above equation, setting the derivative equal to zero, and solving for d_1 . The maximum packing efficiency of the coarse component in such a system is 58.6%.

In order to increase the packing fraction of the first component to about 66%, a second component was added. The size of the second component was selected such that it would be small enough to fit into the large voids in the coarse component matrix and large enough so that it could not migrate through the paths between large voids. The size of the second component was determined to be 0.6 mm, about one-half the size of the coarse component.

Several runs were made in which the two-component first fraction was blended, added to a sample jacket, held in place by a "thimble," and infiltrated with -325 mesh powder. The model runs, made with angular blast-cleaning grit, yielded densities of about 82% TD in which 20% of the material loaded was -325 mesh. The -325 mesh fraction was the stand-in for PuC in the mixed-carbide system.

The model conditions required to yield a vibratorily compacted fuel body of 80% coarse material, 20% finely divided material, and 82% packing fraction were set. At this point the process was ready for application. The charges for the mixed carbides, based upon a 100-g total charge, were: 62.4 g of -12 +14 mesh material, blended with 17.6 g of -30 +35 mesh material, into which was infiltrated 20.0 g of -325 mesh powder.

Preparation of Jackets

All jackets, as received, were inspected, measured, and cleaned before entering the process. The volume of the lower 356 mm of jacket was determined in order to permit calculation of the charge required to obtain a 356-mm-long fuel column. This was required because the jackets made from each material varied in inside diameter. A gauge, precisely 356 mm long, was dropped into the jacket and the retainer rod was adjusted so that the shoulder of the top plug seated on the jacket lip at the same time the retainer plug touched the gauge. After gauging, the jackets were cleaned with acetone and alcohol, and dried at 105°C for not less than one hour.

After cleaning, a stud was screwed into the threaded bottom plug and the jacket was wrapped with a layer of vinyl tape. The stud was used to attach the jacket to the platen of the vibrator used for compaction. The vinyl tape was required to prevent alpha contamination of the jacket exterior. Many schemes to overcome excessive contamination of the jacket were tried; the most successful was the vinyl wrap technique.

The procedure for wrapping the jacket in vinyl tape is described below. The bottom plug and stud body were covered with one 2.5-cm-wide ply of a tape similar to 3M "Scotch" electrical tape. The single turn of tape was applied to mask the irregularities of the end plug and stud. The jacket was then wound to within 10 cm of the top with a single spiral wrap of the same vinyl tape, each revolution overlapping the preceding turn by about 4 mm. The final 10 cm of jacket was wound in a similar manner with 3M "Scotch Magic Tape No. 810." The final wrap of mending tape was necessary because the vinyl tape was too thick to allow the "thimble" to seat against the jacket top. The use of the "thimble" will be described in the Fuel-loading Procedure section, p. 11.

All fuel-element hardware was pouched, flushed with helium, and sealed into vinyl bags before being introduced into the helium-atmosphere gloveboxes of the Plutonium Fabrication Facility at ANL.⁷

Preparation of Mixed Carbide

The materials for the manufacture of the ten mixed-carbide specimens were a combination of purchased and products synthesized at ANL. The uranium carbide, a product of Kerr-McGee Oil Industries, Inc., had the following pertinent properties:

I. Chemical Analysis (in w/o)		
A.	Uranium	95.12
B.	Carbon	4.59
C.	Oxygen	0.47
II. Isotopic Analysis (in w/o) of Total Uranium		
A.	U^{234}	1.05
B.	U^{235}	93.15
C.	U^{236}	0.25
D.	U^{238}	5.56
III. Physical Measurement		
A.	Density (g/cc)	13.28
B.	Percent of Theoretical (UC) Density	97.4

The plutonium carbide was synthesized by the direct reaction of carbon and plutonium, and had the following properties:

I. Chemical Analysis (in w/o)		
A.	Plutonium	95.02
B.	Carbon	4.85
C.	Oxygen	0.044

II. Isotopic Analysis (in w/o) of Total Plutonium

A. Pu ²³⁹	94.99
B. Pu ²⁴⁰	4.61
C. Pu ²⁴¹	0.39
D. Pu ²⁴²	0.02

III. Physical Measurement

A. Density (g/cc)	13.24
B. Percent of Theoretical (PuC) Density	97.4

Massive chunks of uranium monocarbide were reduced to -12 +14 mesh by pounding between a hammer and a steel mortar. This system of particle-size reduction was difficult to control and was soon abandoned in favor of a small Plattner mortar. The product from size reduction was screened and all material finer than 14 mesh was comminuted by Plattner mortar to obtain a -30 +35 mesh fraction. All screening and separation operations were carried out with U. S. Sieve Series screens. Using the Plattner mortar it was possible to accept about 20% of the input in such a narrow sieve range as -12 +14. This method of size reduction is tedious and time-consuming, however. All the plutonium carbide charged to the mixed-carbide specimens was in the form of -325 mesh powder. Powders of this size were obtained by comminution in a micropulverizer.

The fuel charge to an individual tube was calculated on the basis of that particular tube's volume and the achievement of 82.1% void packing fraction. The fuel charges were weighed out, placed in plastic bottles, and stored in a dessicator under vacuum until used. A typical load of mixed carbide for a 12.01-cc column was 81.533 g -12 +14 mesh UC, 23.002 g -30 +35 mesh UC, and 26.134 g -325 mesh PuC.

Preparation of Solid-solution Carbide

The uranium-plutonium carbide for the assembly of the five solid-solution carbide rods was synthesized at ANL by an arc-melting process.⁸ Briefly the process involves the melting of weighed quantities of uranium and plutonium over carbon. The reactants are charged to a water-cooled, electrically grounded, copper hearth and melted by the discharge of an electric arc from a graphite electrode in a low-pressure (0.5 kg/cm²) argon atmosphere.

About 800 g of (U · Pu)C in the form of eight buttons were synthesized by the above method. The buttons, which weighed approximately 100 g, were remelted six to eight times to promote homogeneity. A sample from each button was analyzed for oxygen and carbon content. The oxygen content varied from 42 to 108 ppm, the carbon from 4.42 to 5.51%. The following tabulation is a list of the average properties of the eight carbide buttons.

I. Normalized Chemical Composition (in w/o)		
A.	Uranium	76.33
B.	Plutonium	19.07
C.	Carbon	5.11
D.	Oxygen	0.0066
II. Isotopic Analysis		
A.	Total Uranium (w/o)	
1.	U ²³⁴	1.00
2.	U ²³⁵	93.43
3.	U ²³⁶	0.23
4.	U ²³⁸	5.34
B.	Total Plutonium (w/o)	
1.	Pu ²³⁹	91.38
2.	Pu ²⁴⁰	7.89
3.	Pu ²⁴¹	0.70
4.	Pu ²⁴²	0.03
III. Physical Measurement		
A.	Density (g/cc)	13.21
B.	Percent of Theoretical (0.8U · 0.2Pu)C Density	97.1

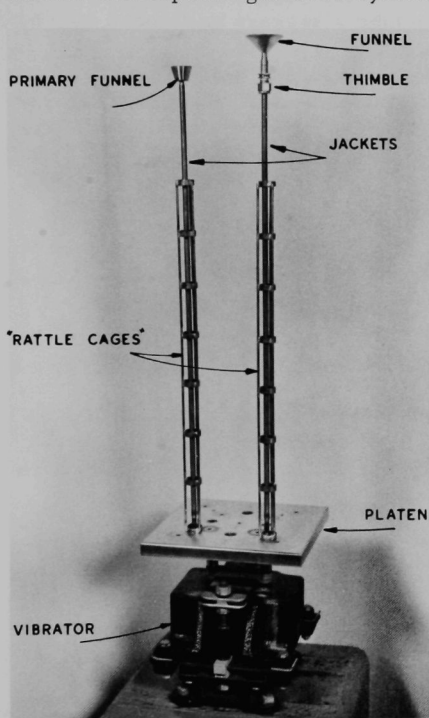
The low yield of -12 +14 and -30 +35 mesh fractions from the size reduction of uranium carbide, previously discussed, and the minimal available quantity of solid-solution carbide caused a supply problem which could best be solved by increasing the yield of desired size fractions. It was noted during the compaction of mixed-carbide rods that considerable attrition of the original size fractions was brought about by the vibrator driving force. The attrition appeared to offer a potential for achieving higher density. An increase in the yield from particle-size reduction was thus accomplished by affecting the attrition before the particles were loaded. Accordingly, three size ranges, -10 +18, -35 +50, and -325, were produced for the compaction of the solid-solution carbides. The Plattner mortar was used to provide the -10 +18 and -35 +50 mesh fractions, and a yield of about 80% of the combined fractions was realized. The -325 mesh fraction was obtained by reduction in a micropulverizer. The fuel charged to each tube was determined on the same basis as for the mixed-carbide system. The -10 +18 fraction was treated as the -12 +14 fraction, and the -35 +50 fraction was substituted for the -30 +35 mesh material; otherwise, all operations up to material storage were identical to those employed in the mixed-carbide preparation.

About 200 g of -50 mesh solid-solution carbide was recovered by consolidation. Consolidation entailed sintering the powder into a loosely held "cinder" and remelting the cinder in the arc process previously described. For the consolidation operation the graphite electrode originally used was replaced by a tungsten electrode. The resulting button was

recrushed, sieved, and analyzed. Analysis showed 75.42% uranium, 19.56% plutonium, 4.89% carbon, 0.15% oxygen, and 0.10% tungsten.

Fuel-loading Procedure

All work on the unjacketed carbide materials was carried out in the helium-atmosphere glovebox system of the ANL Plutonium Fabrication



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Fig. 2. Fuel Jackets with Thimble and Rattle Cage Mounted on Vibrator

Facility. The equipment employed for the compaction of the carbides into metal jackets consisted of an electromagnetic vibrator (Syntron Type V-60) controlled by a rheostat and rectifier for operation at 60 cps. The acceleration impressed upon the specimens during vibration was sensed by an Endevco, Model 2215C accelerometer that was attached to the underside of the vibrator platen. The output from the accelerometer was fed into a Hewlett-Packard, Model 120 B, oscilloscope. The following procedure is the evolutionary product of the simultaneous development of techniques and fuel-specimen manufacture.

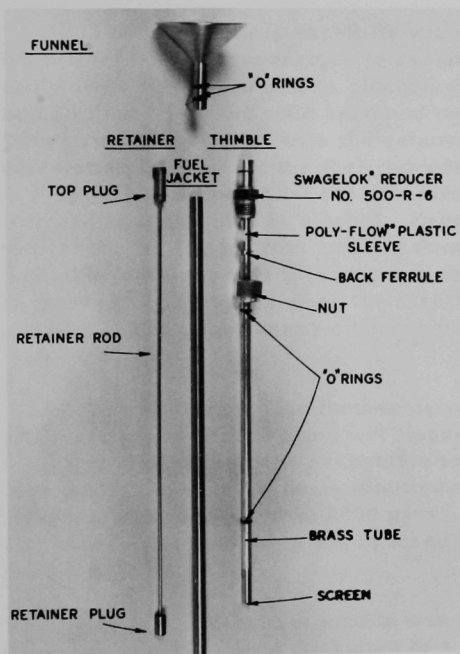
The jackets were attached to the vibrator platen by a stud that was threaded into the bottom plug. Figure 2 shows two fuel tubes, without protective wrapping, mounted on the platen of a vibrator. After the jackets were attached to the platen, a "rattle cage" was dropped over the tube and a funnel was inserted in the open end (tube on left in Fig. 2). The "rattle cage" is a loose-fitting cage fashioned of thin metal rings attached by light-gauge rods. The purpose of the cage

is to limit bridging of the infiltrating fraction by imparting a random secondary mode of vibration normal to the axial vibration. The primary funnel, which was about 22 cm long, was used to direct the fuel charge into the tube and to protect the top, inside surface of the tube wall from excessive contamination.

The preweighed coarse and intermediate-size fuel fractions were mixed thoroughly in a small V-blender. After blending for about 3 min the

charge was poured into the jacket. During the introduction of the blended fuel charge the vibrator was running at its lowest setting, which imparted a force of 5 g. The pouring operation took from 35 to 40 sec, and the fuel level was invariably within 3 mm of final depth. After the introduction of the blended fuel charge, the vibrator was stopped and the primary funnel was carefully removed. The care exercised during removal of the funnel was to prevent the funnel from being scraped along the jacket wall and thereby depositing excessive contamination.

The primary funnel was replaced by a "thimble" and funnel (tube on right in Fig. 2). The construction of a thimble and a fuel retainer is



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*Trade name of Crawford Fitting Co.

**Trade name of Imperial Eastman Corp.

Fig. 3. Details of Thimble and Fuel-column Retainer

shown in Fig. 3. The thimble, which is essentially a restraining device, is made up of a brass tube soldered into a standard "Swagelok" reducer. The bottom of the brass tube is closed by a screen with mesh larger than the most finely divided fuel fraction and smaller than the coarser fractions. The purpose of the screen is to hold the fuel column in place while the finer fraction is being infiltrated into the bed of coarse particles. Squeezing the split plastic sleeve and the back ferrule between the nut and the reducing fitting holds the thimble in place at the top of the fuel jacket and thereby maintains the screen at a fixed depth in the tube. The O-rings prevent the infiltrating fraction from being conveyed up the fuel tube wall and subsequent contamination of the fuel-rod hardware.

The thimble is seated against the fuel jacket top by downward pressure on the reducer while the fuel tube is being driven at 60 cps and 15 g. When the thimble is seated, vibration is halted, and

the nut is tightened over the back ferrule and the split plastic sleeve until the thimble is securely attached to the fuel tube. The funnel is then inserted into the reducer and the tube is ready to receive the fine fuel fraction. The above procedure was employed for the loading of both solid-solution and mixed-carbide elements.

The addition of the fine fraction was made through the thimble. In the case of the mixed-carbide system the fine fraction consisted of a pre-weighed quantity of PuC. About 4 hr of vibration at 15 g were required to infiltrate the coarse-fraction matrix with the -325 mesh PuC.

In the solid-solution case an attempt was made to reach maximum density in the fuel body. The -325 mesh fraction was added to the matrix as long as it would accept it. This approach required addition of the fine fraction for up to 8 hr while vibrating the tube at 60 cps and 15 g.

Assembly of Fuel Rod

The thimble was carefully withdrawn and the vinyl tape was stripped from the fuel rod preparatory to assembly. The fuel specimens were partially decontaminated before closure welding to prevent plutonium from being alloyed into the weld bead and permanently contaminating the surface. Decontamination began with the removal of the 3M "Scotch Magic Tape" by dissolution in acetone; the fuel rod was then wiped clean with dry tissues. A 1-cm strip of the jacket top was cleaned, both inside and out, by wipes with swabs moistened in alcohol. Excess alcohol was wrung from the swab to prevent the liquid from running down the jacket wall and into the fuel bed. The retainer was inserted into the fuel tube and the tube transferred to the welding glovebox when the direct count on the swabs was below 200 dpm. The preliminary cleaning just described usually required less than 24 wipes.

In the welding line further decontamination was accomplished by alternate wipes with dry and wet tissue. The dried tissues were counted directly. The jacket was acceptable to welding when such surveys showed less than 10 dpm loose contamination and direct count of the jacket-retainer interface showed less than 2000 dpm. The closure of a decontaminated rod was made by a girth weld which fused the top plug of the retainer to the fuel jacket.

The final decontamination was carried out in an air-filled glovebox. Cleaning of the fuel rod consisted of a wipe with a dry tissue which was counted to determine the extent of contamination. The specimen was then wiped with alcohol and No. 400 grit "wet or dry" paper. A wipe with tissue, wet with alcohol, followed the treatment with 400 grit paper. After the wet wipe with tissue a wipe for counting was taken. The process of alternate cleaning with wet grit paper, wet tissue wipes, and samples for counting was repeated until a count of less than 10 dpm was found on the complete tube-surface wipe.

The rod from the above cleaning treatment was taken out of the glovebox and direct alpha surveys were made. If the readings were in excess of 500 dpm, the alternate treatment with wet grit paper and wet

tissue wipes was repeated until a direct count less than 500 dpm at any point on the rod was realized. The direct count was taken on a 5-cm-wide strip around the fuel rod. The entire fuel rod length was surveyed in this manner.

Evaluation of Fuel Rod

The evaluation of the fuel specimens included contamination survey, leak detection, X radiography, gamma radiography, autoradiography, and some eddy-current examination. Completed decontaminated specimens were surveyed directly with a gas proportional counter for "fixed" contamination and by the counting of dry wipes for "loose" contamination. In all cases the loose contamination was less than a total of 10 dpm for wipes over the entire rod surface. Except for isolated cases of contaminated welds, direct survey of the rods showed levels of contamination less than 500 dpm.

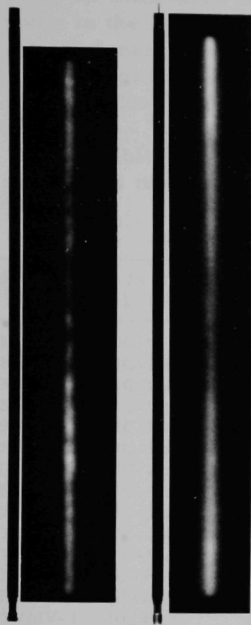
A helium mass-spectrometer leak test at room temperature was made on all fuel specimens. The rod was placed in a chamber connected to a vacuum pump that drew a vacuum around the fuel-rod exterior. When a satisfactory vacuum was attained, the chamber was isolated from the vacuum pump and opened to the leak detector. An acceptable specimen showed a leak rate less than 2×10^{-6} std cc/sec.

After cleaning and leak checking, the specimens were radiographed to determine the distribution of the carbides in the fuel column. The fact that eight different jacketing materials and two fuel systems were present in the batch of specimens manufactured made radiography a challenge. The gamma emission of plutonium isotopes, or their products of decay, was used to indicate the plutonium distribution in the rod by autoradiography. Gamma radiography was carried out, using a cesium-190 source, to show the particle distribution in the several rods. X radiography, as performed on the specimens described, yielded a description of the boundary between the jacket and the fuel particles. Eddy-current techniques were employed to indicate density variation with rod length. Although it was possible to correlate the output from eddy-current examination with the radiographs of the specimens, the eddy-current technique was considered inadequate and in need of development.

RESULTS

Ten specimens of mixed UC and PuC were assembled by the methods discussed. The calculated fuel charges should have resulted in homogeneous specimens of 80% TD; however, X radiography of the fuel columns showed void areas at the tube wall near the retainer. The voids were indicative of low density resulting from the absence of the fine fraction.

This condition was confirmed by autoradiography, which showed an absence of the -325 mesh PuC fraction at the top of the fuel column. The left photograph in Fig. 4 is an X radiograph and autoradiograph of a fuel rod showing this condition. The low density at the top of the fuel column was brought about by the deviation of the carbide system from the model system discussed in the Process Development section, p. 6.



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Fig. 4

Autoradiographs and X Radiographs of Typical Mixed and Solid-solution Carbide Fuel Specimens

The model system was made up of angular-shaped particles of malleable material. The effect of the highly brittle nature of the carbide fuel was to provide a wider range of particle sizes during settling of the matrix than was introduced into the jacket as feed. Experience gained with some rejected specimens showed that 20% of a carbide fraction could be reduced to a smaller fraction during loading of the matrix material alone. Since the loading of the -325 mesh fraction took as much as 8 hr, a considerable attrition could be expected. Although one would not expect the generation of additional void volume by particle cleavage, a similar effect does exist.

We have found² that the packing efficiency of regular tetragonal shapes may be increased by rounding the edges of the particles. The increase in packing efficiency of milled particles is brought about by at least three conditions: (1) the rounding of edges reduces surface-to-volume ratio (the particle approaches spherical dimensions); (2) the voids in the matrix have rounded walls which reduce resistance to the entrance of the fine fraction; (3) extremely fine powders resulting from attrition add

another size fraction to the particle-size distribution. Since the extent to which the carbides would be affected by abrasion was not known, it was difficult to predict the degree of this effect.

Five specimens of solid-solution (U·Pu)C were assembled in which maximum uniform density was desired. The void-packing efficiency reached was $(86.7 \pm 1.5)\%$ for 97.5% dense particles. An indication of the uniformity of the fuel column may be seen in the photograph on the right side of Fig. 4, which is an X radiograph and autoradiograph of a solid-solution fuel column in vanadium jacket. This figure shows the increase in uniformity of plutonium distribution inherent in solid-solution feed materials.

One rod containing mixed carbide was made on the basis of information gleaned from the solid-solution specimens. The -325 mesh fraction, made up of 18.4% UC and 81.6% PuC, was added until no additional material could be infiltrated into the UC matrix. The finished fuel rod showed no voids in the X radiograph. The total packing fraction was 86.0%, the theoretical density was 83.8%, and the plutonium carbide content was 19.3%. Autoradiograph revealed an area approximately 3 cm in the center of the rod to be low in plutonium content.

Table II is a compilation of the content of each of the sixteen fuel specimens made.

TABLE II. Properties of Fuel Specimens

Specimen No.	Fuel Form	Packing Fraction, %	Theoretical Density, %	UC, g	PuC, g	(U · Pu)C, g	U, g	Pu, g
NMV-3	UC-PuC	82.1	80.0	107.264	26.318		102.0	25.0
NMV-4	UC-PuC	82.1	80.0	104.541	26.137		99.4	24.8
NMV-7	UC-PuC	82.1	80.0	105.874	26.469		100.7	25.2
SMV-1	UC-PuC	82.1	80.0	104.781	26.197		99.7	24.9
SMV-2	UC-PuC	86.0	83.8	107.644	25.706		102.4	24.4
HMV-1	UC-PuC	82.1	80.0	111.621	27.908		106.2	26.5
HMV-2	UC-PuC	82.1	80.0	111.621	27.911		106.2	26.5
HMV-4	UC-PuC	82.1	80.0	111.609	27.906		106.2	26.5
HMV-5	UC-PuC	82.1	80.0	111.629	27.913		106.2	26.5
TMV-2	UC-PuC	82.1	80.0	104.901	26.227		99.8	24.9
TMV-3	UC-PuC	82.1	80.0	104.904	26.229		99.8	24.9
NMV-11	(U · Pu)C	85.1	82.9			137.599	105.0	26.2
NMV-12	(U · Pu)C	87.8	85.5			138.022	105.4	26.3
TVMV-1	(U · Pu)C	85.9	83.7			134.714	102.8	25.7
HWMV-1	(U · Pu)C	85.1	82.9			137.366	104.9	26.2
VMV-1	(U · Pu)C	88.2	85.9			137.843	105.2	26.3

RECOMMENDATIONS

The hand operation for particle-size reduction and control was time-consuming or wasteful of material. To alleviate this condition a crusher and a study of crushing characteristics of brittle materials is needed to determine optimum comminution conditions.

Although the fuel specimens manufactured were free of loose contamination, the effort applied to decontamination was considerable. Techniques and methods by which contamination during fabrication of fuel specimens can be controlled are required. The philosophy of never allowing an outer surface to become contaminated is an acceptable one, but satisfactory cleaning techniques are necessary when practice falls short of theory.

The third operation most demanding of time was the infiltration of the fine component into the matrix of coarser particles. Very little has been done to determine the conditions influencing the rate at which a fine fraction will enter a bed of particles. A study of such a nature is necessary to determine the parameters controlling production rate.

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REFERENCES

1. J. E. Ayer and F. E. Soppet, Vibratory Compaction: I. Compaction of Spherical Shapes, J. Am. Ceram. Soc 48(4), 180-183 (1965).
2. J. E. Ayer and F. E. Soppet, Vibratory Compaction: II. Compaction of Angular Shapes, J. Am. Ceram. Soc., in press.
3. W. G. Tope, Production of Plutonium Monocarbide by Reaction of Elemental Plutonium and Carbon, ANL-6969 (July 1965).
4. J. E. Ayer and F. E. Soppet, Method and Apparatus for Vibratory Compaction, U. S. Patent applied for.
5. J. J. Hauth, Vibrationally Compacted Ceramic Fuels, HW-67777, (Jan. 10, 1961).
6. S. W. Porembka, C. B. Boyer, and J. J. Hauth, Fabrication of UO₂ Fuel Rods by Vibrational Compaction and Pressure Bonding, BMI-1637 (June 28, 1963).
7. A. B. Shuck and R. M. Mayfield, The Process Equipment and Protective Enclosures Designed for the Fuel Fabrication Facility No. 350, ANL-5499 (Jan 1956).
8. O. L. Kruger, Preparation and Some Properties of Arc-cast Plutonium Monocarbide, J. Nuclear Materials 7(2), 142-150 (1962).

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